



TITLE:

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PAPER IV

Colloid Morphological and Crystalline Studies of "Bikini Dust" fallen on the No. 5 Fukuryu Maru by Electron Microscopy and Diffraction Methods.

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INTRODUCTION

"Bikini dust" fallen on the No. 5 Fukuryu Maru on the 1st of March, 1954, by the H-bomb test of U.S.A. at Bikini Atoll has been studied from the radiochemical point of view by many investigators. An attempt has been made in this investigation to observe the shape and the size of the dust particles by electron microscopy from the colloid morphological point of view and further to study the crystal type by means of electron diffraction method. That is to say, the authors pursued whether the "Bikini dust" was the broken pieces of coral reef or the volatilized matter by the explosion. If the fission product particles were found, it was expected to be analyzed, but they were not found. X-ray analysis was also applied to analyze the crystals.

The samples used in this investigation were as follows:*

1. Cotton wiped over the part having the strongest radioactivity on the deck of the No. 5 Fukuryu Maru.
2. Dust adhered on fins of fishes.
3. Dust fallen on the No. 5 Fukuryu Maru, which had been collected by the fishermen and kept at Shizuoka University.

EXPERIMENTS AND RESULTS

A. Dusts wiped by cotton and adhered on a fin

1. Disposition of samples

The cotton, with which the part having the strongest radioactivity of the No. 5 Fukuryu Maru had been wiped, was black dirty pieces. From about 0.5g of the cotton the dust was washed out by stirring with a glass rod in the test tube with 10 cc of water. The prepared suspension was mounted on specimen holders for electron microscopy and was dried. The radioactivity of each specimen registered 150~250 counts per minute at the distance of 1 cm from the mica window of G-M tube (March, 27).

White fine particles taken from a fin of a tuna were pulverized by the tip

* On this occasion we wish to express our thanks to Prof. T. Shiokawa, Shizuoka University, and Dr. T. Maekawa, Chief of Sanitation Division of Shizuoka Prefecture, for their kindness to offer the sample.

of a platinum wire with a drop of water in a small basin, and the suspension was mounted on specimen holders as usual. Each specimen registered 100~250 counts per minute. (March, 30)

2. Results of the electron microscopic examination

The sample made from the cotton was almost rubbish which has not distinct shape as shown in Photo. 1 and a few sphere particles, which were a few μ in size and seemed to be carbon particles, were contained as shown in Photo. 2. In the particles sampled from the fin, many spherical particles, about 1 μ in size, were found as shown in Photos. 3, 4, and 5. But the constitution of the spherical particles could not be determined by the electron diffraction.

B. Studies on "Bikini Dust"

1. Observation by optical microscope

"Bikini dust" is white granules. They have not regular shape as shown in Photo. 6. All of 165 granules given to the authors were photographed by an optical microscope and the distribution of particle size (Green size) was obtained as shown in Fig. 1.

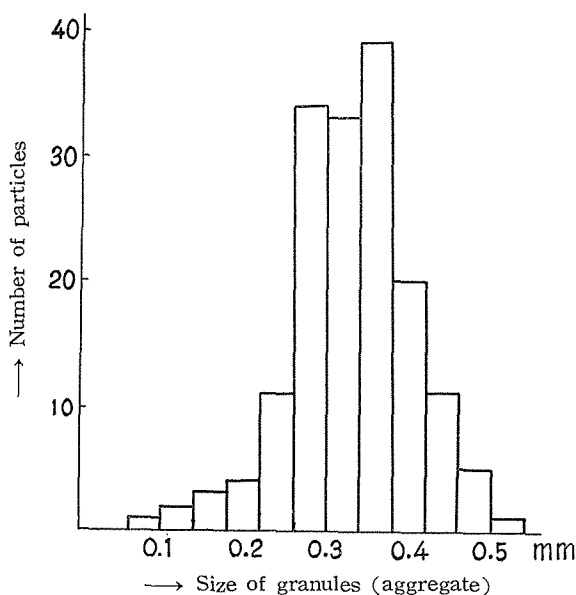


Fig. 1. Distribution of particle size (aggregate) of "Bikini dust" granules by optical microscope.

From the size distribution the mean volume diameter of granules $(\sum nd^3 / \sum n)^{1/3}$ was calculated as 0.321 mm. Total weight of the dust (165 granules) was 6.4 mg and the mean weight of one granule was 0.039 mg. Then the apparent specific gravity of the dust granules became 2.42. This value is somewhat less than the

specific gravity of CaCO_3 (Calcite 2.71, Aragonite 2.88). 105 granules of the dust registered 1800 counts per minute at the distance of 2 cm from the mica window of G-M tube (July, 6).

2. Morphological studies by electron microscopy

A few granules of "Bikini dust" were taken in a small basin and pulverized by the tip of a platinum wire with a drop of water. The suspension was mounted on specimen holders for the electron microscopy as usual. The specimens registered 30~150 counts per minute at the distance of 1 cm from the mica window of G-M tube (May, 12).

85 electron micrographs were taken. Some of them are shown in Photos. 7~16. Unit particles of the dust have definite shapes as cube (Photos. 7, 8 and 9) and spindle (Photos. 10 and 11). Aggregates of these unit particles, such as sphere or confetto (Photos. 12, 13 and 14), are also found. Fine particles which have not definite shape (Photos. 15 and 16) are found, too. The size distribution curves for the cubic and spindle particles are indicated in Fig. 2. The relations of the shape to

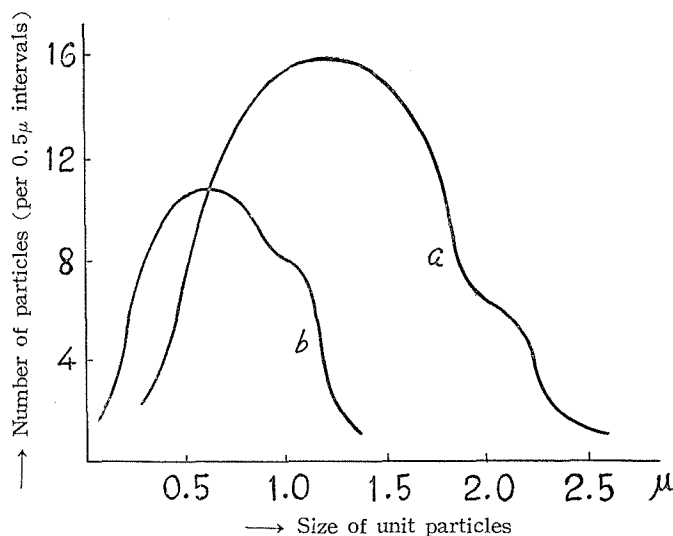


Fig. 2. Distribution of unit particle size of "Bikini dust" by electron microscope.

a : cubic particles

b : spindle particles

Table 1. The shape and the size of unit particles

Shape	Size (mean)
Cube	1.2 μ
Spindle	0.65
Sphere (confetto)	1.1
Fine particles	<0.1

the mean size are shown in Table 1.

To compare with "Bikini dust", three kinds of coral reef (Tinian, Kuseie and one another islands in South Pacific) were observed by the electron microscope after powdering by an agate mortar. The particles which construct the coral reef have not regular shape as shown in Photos. 17 and 18.

3. Crystal structure of unit particles studied by electron micro-diffraction method

The selected area electron diffraction technique, the electron micro-diffraction method¹⁾, by means of the three stage electron microscope (SM-C3) was applied to

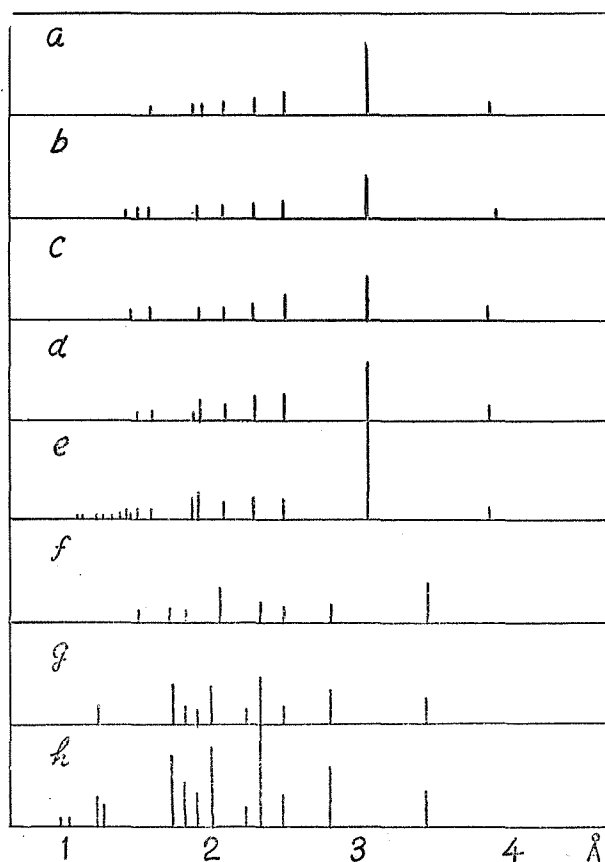


Fig. 3. Diagrams of electron and X-ray diffractions.

- a* : electron diffraction diagram of fine particles.
- b* : electron diffraction diagram of cubic particles.
- c* : electron diffraction diagram of spindle particles.
- d* : X-ray diffraction diagram of "Bikini dust".
- e* : X-ray diffraction diagram of standard calcite.
- f* : electron diffraction diagram of pulverized coral reef.
- g* : X-ray diffraction diagram of coral reef.
- h* : X-ray diffraction diagram of standard aragonite.

study the chemical composition and the crystal structure of "Bikini dust". That is to say, the cubic, spindle and fine particles, which were unit particles of the dust, were studied by the electron micro-diffraction method, and the electron micrographs and the diffraction patterns of their circled positions are shown in Photos. 19, 20, 21 and 22. The electron diffraction pattern of the fine particles gave Debye-Scherrer rings as shown in Photos. 21 and 22. The cubic and spindle particles gave coarse rings as shown in Photos. 19 and 20, and the spacings could be also calculated. The spacings of these crystals were calculated from the diffraction patterns and were shown in Fig. 3 (a), (b) and (c). These diagrams were in agreement with the standard diffraction diagrams of calcite (e). Some other electron micro-diffractions of various particles of "Bikini dust" were taken, but the diffraction pattern which did not agree with calcite was not found.

The selected area electron diffraction technique was also applied to the pulverized coral reef, whose diffraction pattern was shown in Photo. 23. The spacing calculated from the diffraction pattern (Fig. 3 (f)) agreed with that of aragonite (Fig. 3 (h)).

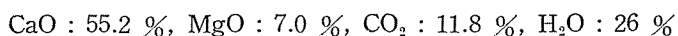
4. Crystal structure studied by X-ray analysis

X-ray analysis was applied to confirm the above results obtained by the electron diffraction. The X-ray diffraction patterns taken from "Bikini dust" (2~3 granules were used) and coral reef are shown in Photo. 24. The spacings calculated from these patterns shown in Fig. 3 (d) and (g) agree with those of calcite and aragonite.

DISCUSSION

"Bikini dust" fallen on the No. 5 Fukuryu Maru is white granules which are about 0.3 mm in diameter and the apparent specific gravity of them is little less than the specific gravity of CaCO_3 . The granules are pulverized easily and give unit particles which have definite shapes such as cube and spindle. The dust is the aggregates of these crystalline particles. The shapes of the unit particles of CaCO_3 were almost cubic or spindle by the authors' observation on various kind of CaCO_3 by electron microscope²⁾. These crystals produced by the liquid reaction are calcite and the shapes agree with the unit particles of "Bikini dust". "Bikini dust" were also decided to be calcite by the electron and X-ray diffractions as described above.

The chemical composition of "Bikini dust" by the investigation of K. Kimura and co-investigators³⁾ are as follows:



According to these data the quantity of MgO is relatively great. But the crystals to be seemed as MgO or MgCO_3 were not found. MgCO_3 was probably mixed into CaCO_3 and crystallized as calcite.

It has been reported⁴⁾ that the temperature of sea water, in which the coral reef

grow, is warmer, the higher the aragonite : calcite ratio is, and that the coral reef in the South Pacific is composed 100 % aragonite. In the authors' experiment, three kinds of coral reef in South Pacific were also decided to be aragonite by the electron and X-ray diffractions.

The coral reef is the crystal of aragonite and "Bikini dust" is calcite. Then "Bikini dust" is decided not to be simply pulverized material of coral reef. Aragonite seemed to change to the stable calcite by thermal transition. The coral reef has not a definite shape, but the unit particles of "Bikini dust" have cubic and spindle shapes that were specially for calcite. Then it was suggested that "Bikini dust" was produced after recrystallization of the coral reef.

It seems that by the H-bomb explosion the coral reef was blown away and just then by a violent fever CaCO_3 (aragonite) was not only decomposed to CaO (decomposition temperature, ca. 900°C), but CaO was melted (melting point, 2572°C) and evaporated (boiling point, 2850°C), and further decomposed to the atomic state, then CaO and Ca crystallized to CaCO_3 (calcite) with H_2O and CO_2 in air. The unit particles were $0.1\sim 3\ \mu$ in size and aggregates of these unit particles were carried by the wind to the No. 5 Fukuryu Maru as the uniform size granules. It seemed that these CaCO_3 particles adsorbed the fission products —probably atomic state— and the fission product particles were not properly found by the electron microscopy and the micro-diffraction method.

SUMMARY

1. "Bikini dust" fallen on the No. 5 Fukuryu Maru by the H-bomb test of U. S.A. on the 1st of March, 1954, is white granules, about 0.3 mm in size, and 2.42 in apparent specific gravity.

2. These granules were composed of unit particles which were cubic or spindle shapes of $0.1\sim 3\ \mu$ in size.

3. "Bikini dust" was decided as calcite by electron micro-diffraction and X-ray diffraction studies. The coral reef is aragonite.

4. It seemed that by the H-bomb explosion the coral reef was evaporated, then it recrystallized to calcite and the aggregates of the unit particles fell on the No. 5 Fukuryu Maru as "Bikini dust".

ACKNOWLEDGEMENT

It is a pleasure to acknowledge the support by the special research grant from the Ministry of Education.

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- 2) E. Suito, M. Arakawa and H. Hasegawa, *J. Soc. Rubber Ind. Japan*, **26**, 216 (1953); E. Suito, *Bull. Inst. Chem. Res. Kyoto Univ.*, **18**, 115, 117 (1949) ; **31**, 69 (1953).
- 3) K. Kimura etc., *Japan Analyst*, **3**, 335 (1954).
- 4) H. A. Lowenstam, *J. Geology*, **62**, 284 (1954) ; cf. H. S. Ladd, J. I. Tracey, J. W. Wells and K. O. Emery, *ibid.*, **58**, 410 (1950)

Electron micrographs

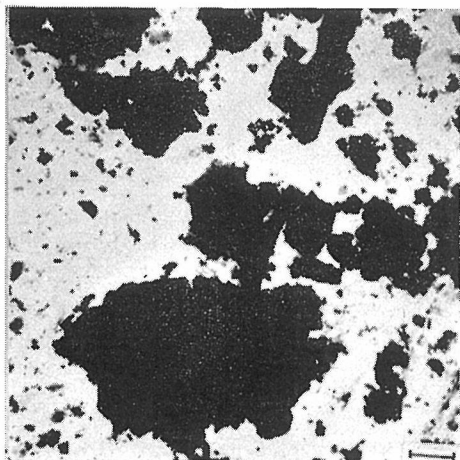


Photo. 1. Dust from the cotton
 $\times 6,000$

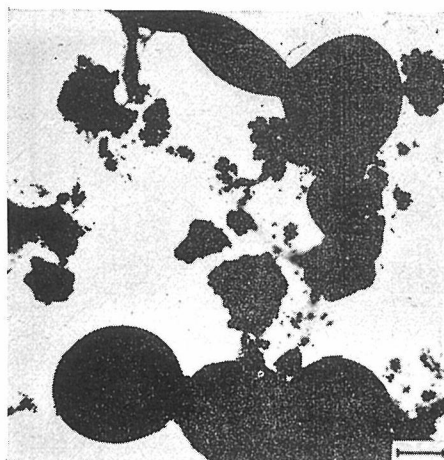


Photo. 2. Dust from the cotton
 $\times 6,000$

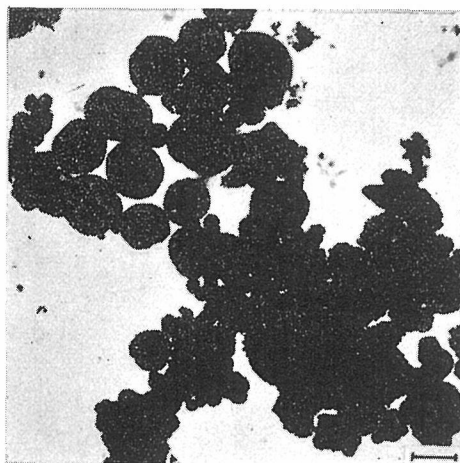


Photo. 3. Dust from a fin
 $\times 6,000$

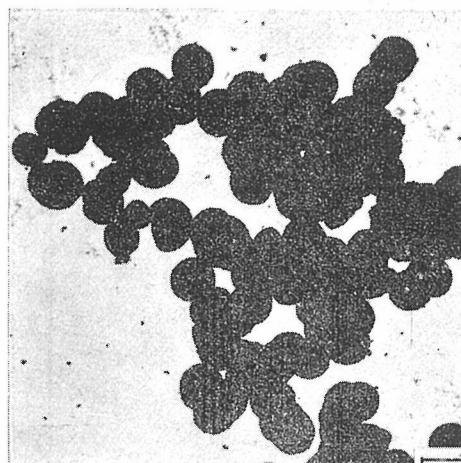


Photo. 4. Dust from a fin
 $\times 6,000$

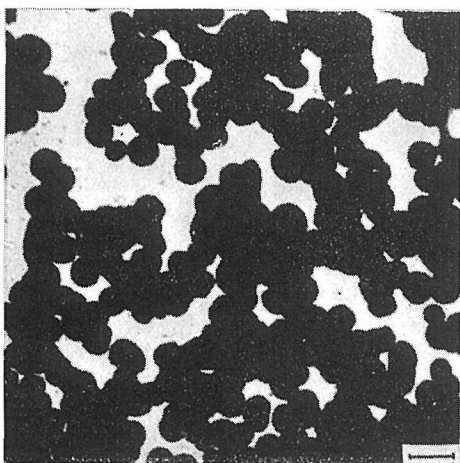


Photo. 5. Dust from a fin
 $\times 6,000$

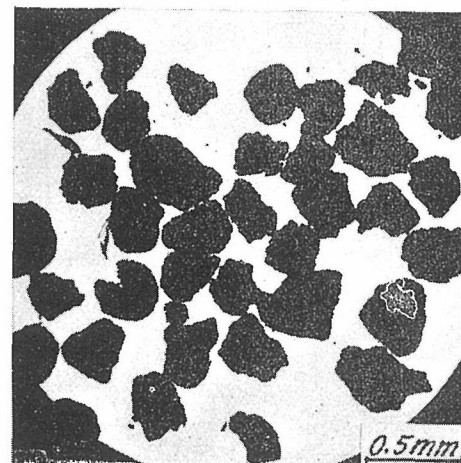


Photo. 6. "Bikini dust" (Optical
micrograph) $\times 25$

Electron micrographs

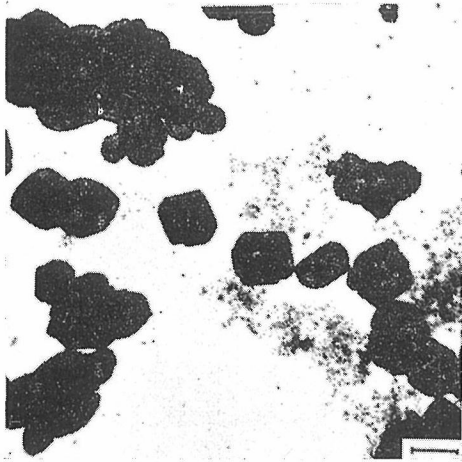


Photo. 7. Bikini dust $\times 6,000$

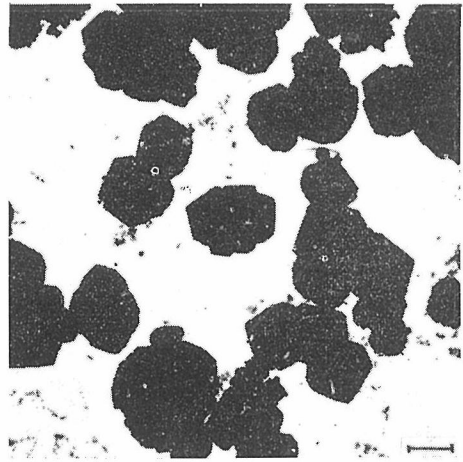


Photo. 8. Bikini dust $\times 6,000$

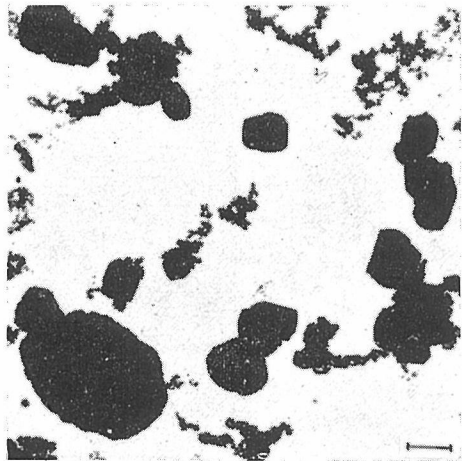


Photo. 9. Bikini dust $\times 6,000$

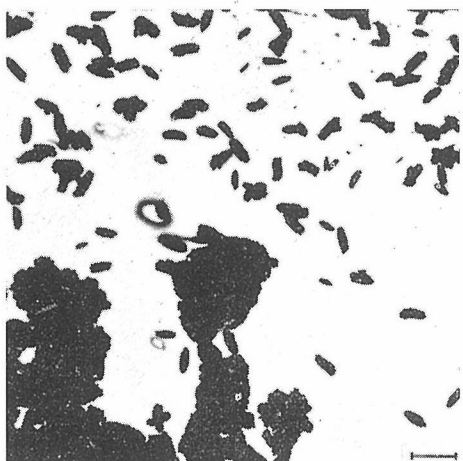


Photo. 10. Bikini dust $\times 6,000$

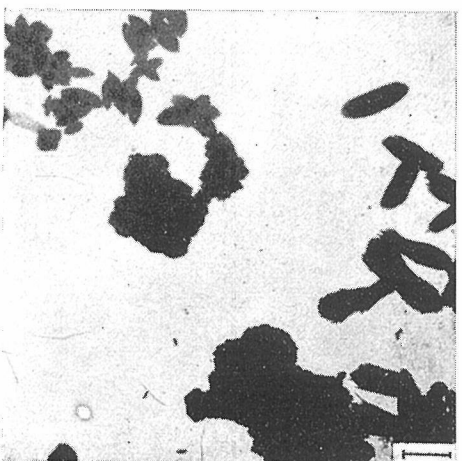


Photo. 11. Bikini dust $\times 6,000$

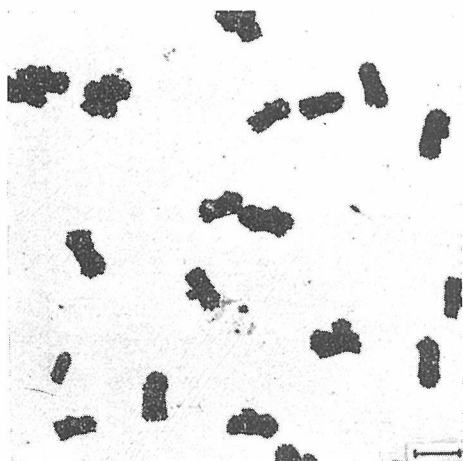


Photo. 12. Bikini dust $\times 6,000$

Electron micrographs

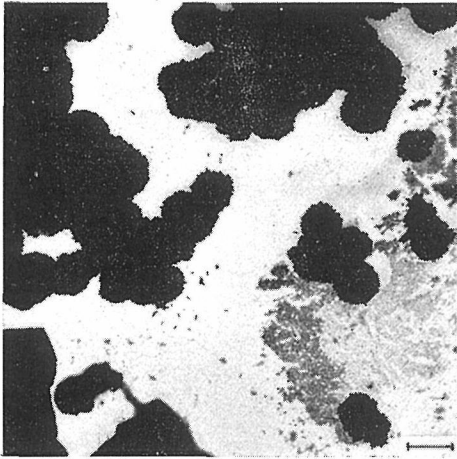


Photo. 13. Bikini dust $\times 6,000$

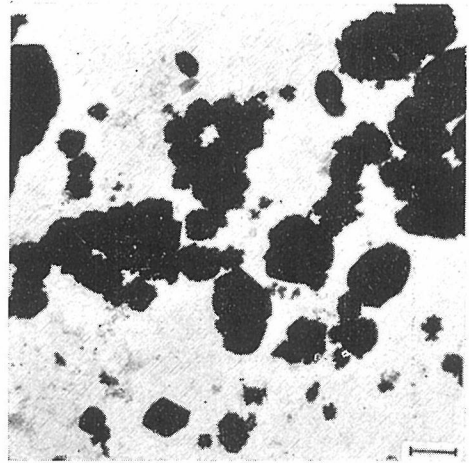


Photo. 14. Bikini dust $\times 6,000$

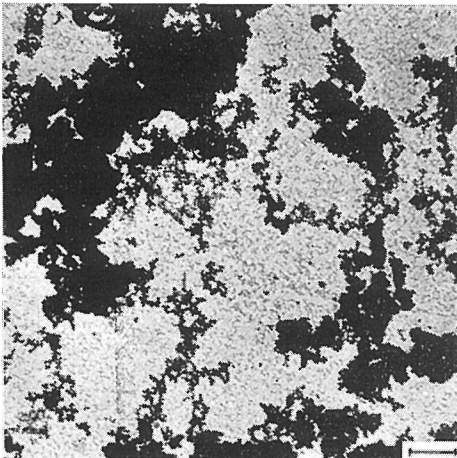


Photo. 15. Bikini dust $\times 6,000$

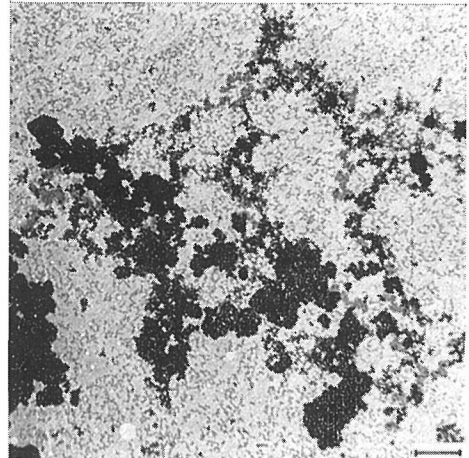


Photo. 16. Bikini dust $\times 6,000$

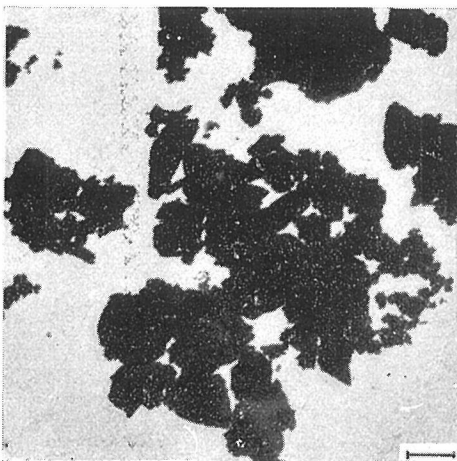


Photo. 17. Pulverized coral reef
 $\times 6,000$

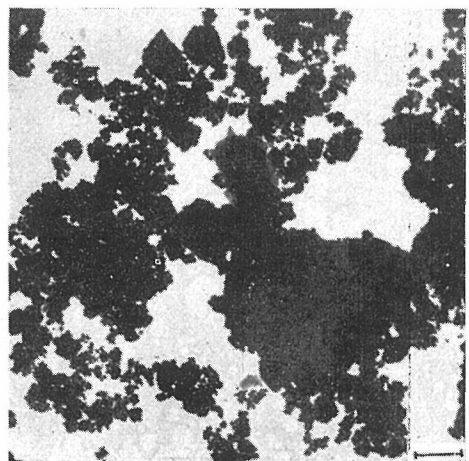


Photo. 18. Pulverized coral reef
 $\times 6,000$

Electron micro-diffraction and X-ray diffraction diagrams

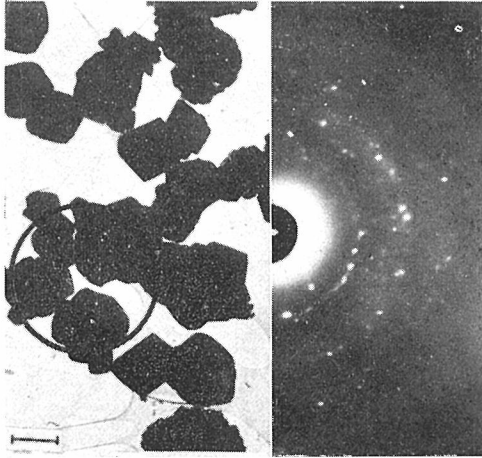


Photo. 19. Bikini dust $\times 6,000$

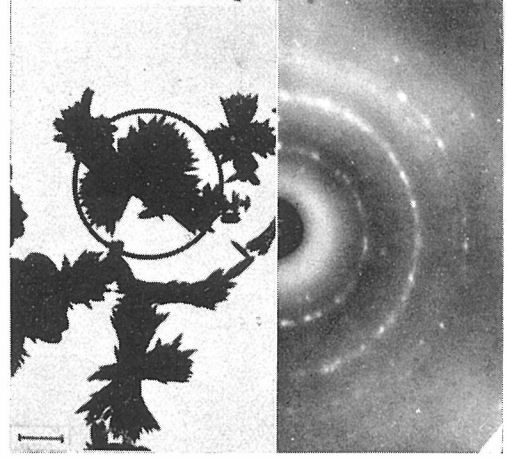


Photo. 20. Bikini dust $\times 6,000$

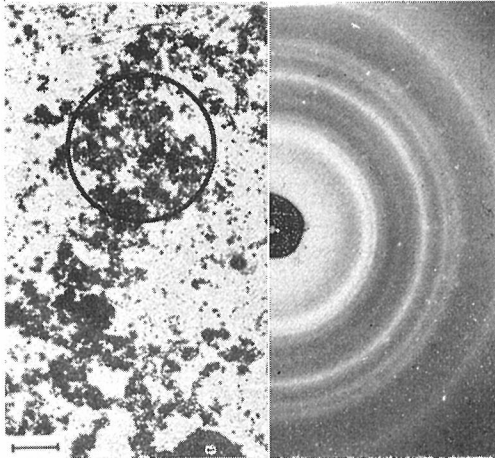


Photo. 21. Bikini dust $\times 6,000$

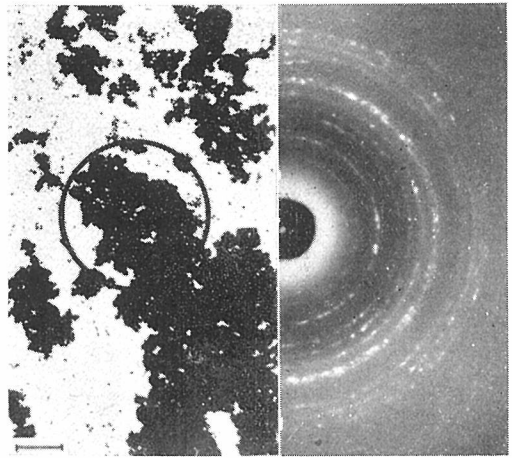


Photo. 22. Bikini dust $\times 6,000$

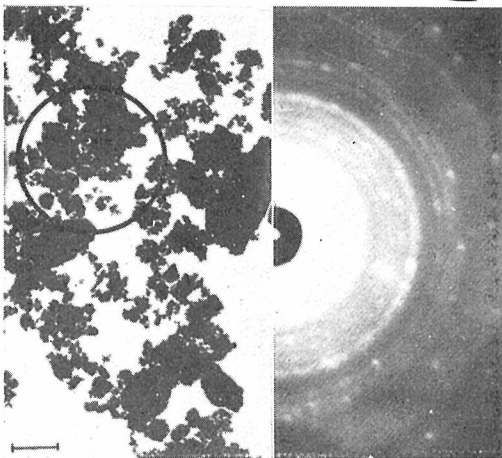


Photo. 23. Pulverized coral reef
 $\times 6,000$

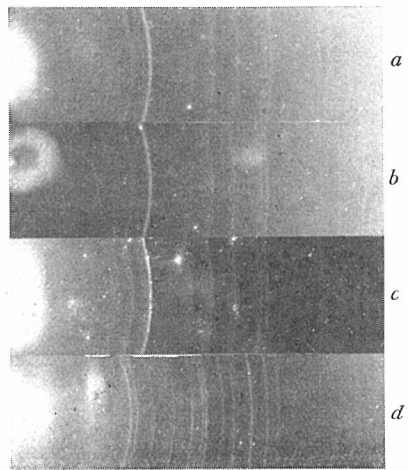


Photo. 24. X-ray diffraction diagrams
a, b : Bikini dust *c* : Standard calcite
d : Coral reef